LOCAL CONNECTIVITY APPROACH TO MICROEMULSIONS : PROOF OF A BICONTINUOUS PHASE IN A LATTICE MODEL(Session III : Complex Fluids, The 1st Tohwa University International Meeting on Statistical Physics Theories, Experiments and Computer Simulations)

AUTHOR(S):

UENO, Yohtaro

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Yohtaro UENO

Dep. of Physics, Tokyo Institute of Technology, Oh-okayama, Meguro, Tokyo 152

"Oil and water" is a synonym of being incompatible. However, when amphiphiles (or surfactants) are added, oil and water become solvent to each other to some extent depending on the relative ratios of three substances. It is because surfactants tend to concentrate as a monolayer at an interface between oil and water, reducing the interfacial tension remarkably. Our main interest is in the single fluid phase of the microemulsions, which is seen for a constant temperature in the low to intermediate region of surfactant concentration c_s and in a broad range of water (or oil) concentraion c_w (or c_o).[1] The bi-continuous microemulsion (bi-ME) exists in $c_w \sim c_o$, with an isotropic percolated sheet of surfactants sitting at the water-oil interface. The mono-continuous MEs (mono-ME) exist in both sides of the bi-ME.

The present study addresses the following questions.

(a) Does the bi-ME smoothly change to the disordered phase (DP) without any singularity, say, as T is raised? In the DP, surfactants are released from the w-o interfaces into the sea of water and oil, whereas in the bi-ME surfactants get together at interface spontaneously into an isotropic percolated sheet.

(b) The same question arises for the change between the mono-ME and bi-ME, when c_w is increased, fixing c_s. The surfactant sheets at interface are closed and of finite sizes in the mono-ME, while they are open and percolated with finite density in the bi-ME.

In some kind of descrete spin systems in $d = 3$, we have rigorously proved the existence of a novel type of ordered phases which closely resemble the bi-ME, by using a new theoretical method.[2] With the help of a Monte Carlo study,[3] we have also obtained the existence of a topological transition in $d = 3$. The topological phase transition without symmetry breaking has been obtained theoretically for the first time, but it is far from full understanding and experimental study is required. For this purpose, it is probably much easier to study the ME system than the spin system, though it might be very difficult to observe singularities of the topological transition in $d = 3$ as suggested from the $d = 2$ case, that is, the Kosterlitz-Thouless transition.

Using Alexander's lattice model[4], we apply the local connectivity approach[2] to it. In the model, water and oil molecules sit on a lattice site. $S_i = 1/2$ (water), -1/2 (oil). Surfactants lie only between NN sites. $\tau_{ij} = 1$ (present), 0 (absent). We neglect interaction between surfactants for simplicity, The interaction energies $(\varepsilon_1, \varepsilon_2, \varepsilon_3)$ are defined as in the partition function, assuming $\varepsilon_3 > \varepsilon_2 > \varepsilon_1$:

$$Z(T, H, \{\varepsilon_i\}) = \sum_{\{S_i\}} \sum_{\{\tau_{ij}\}} \prod_i e^{2\beta H S_i} \prod_{<ij>} e^{-\beta V_{ij}},$$

where

$$e^{-\beta V_{ij}} = (1-e^{-\beta\varepsilon_3})\delta(\hat{S},0)\delta(\tau,0) + (e^{-\beta\varepsilon_1} - e^{-\beta\varepsilon_3})\delta(\hat{S},1)\delta(\tau,1) + (e^{-\beta\varepsilon_2} - e^{-\beta\varepsilon_3})\delta(\hat{S},0)\delta(\tau,1) + e^{-\beta\varepsilon_3}.$$

with $\hat{S}_{ij} = |S_i - S_j| = 0$ or 1. The chemical potential for surfactants is included in ε_1 and ε_2. In obtaining the equation, we have made use of $\sum_{k=0,1}\sum_{\ell=0,1}\delta(\hat{S},k)\delta(\tau,\ell) = 1$.

According to the theory of local connectivity[2] which defines the physically connectivity that leads to phase transitions, the last term corresponds to *disconnection* with probability $e^{-\beta\varepsilon_3}$, while the others represent different kinds of *physical connections* with the respective probabilities. For example, the first term connects the same kind of molecules and the second does w- and o-molecules via a surfactant to form a surfactant sheet at interface.

First we study the physically connected model that has no disconnection ($e^{-\beta\varepsilon_3} = 0$). We prove that it has a bi-ME at high temperatures. Then we examine whether the ME is stable or not at the presence of disconnection ($\varepsilon_3 < \infty$).

The following are the results we have obtained. Details will be given elsewhere.

(i) There is a topological phase transition from the DP to the bi-ME in some parameter range, where an isotropic percolated sheet forms spontaneously. The order parameter is the area of the percolated sheet per unit volume (or the number of surfactants of it).

(ii) There is another topological phase transition from the mono-ME to the bi-ME, where closed sheets get together into a percolated one.

It is also suggested that there is a topological phase transition from the mono-ME to the DP, where the area of spontaneous intefaces (i.e., micelles) vanishes. The order parameter is the density of micelles (or the surface area of them).

We note that the connectivity treated by some authors (see the latter of ref.1)) is not physical connection but geometrical one.

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2) Y. Ueno, *J. Stat. Phys.* **80**, 841 (1995).
3) Y. Ueno and K. Kasono, *Phys. Rev.* B**48**, 16471 (1993).
4) S. Alexander, *J. Physique Lett.* **39** (1995) L1.